

THEORETICAL AND MATHEMATICAL PHYSICS

Selection of Stable Growth Conditions for the Parabolic Dendrite Tip in Crystallization of Multicomponent Melts

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Received February 3, 2012

Abstract—Free growth of a dendrite crystal in a stationary multicomponent melt is investigated. A mathematical model of the process is developed and its analytic solution is constructed. A stability criterion is obtained for a 2D parabolic dendrite and the refined relation is derived for the growth rate of the dendrite tip taking into account the anisotropy of surface tension at the solid phase–melt interface. It is shown that the accumulation of impurity in the front of a dendrite makes it tip thinner and reduces its velocity. The theory contains limiting transitions to the familiar dendrite growth criteria for one-component and binary systems.

DOI: 10.1134/S106378421303002X

INTRODUCTION

It is well known that solidification processes play an important role in metallurgy and completely determine the physical and mechanical properties of crystallized substances in many cases. Mathematical models of such processes originate from the classical Stefan formulation describing crystallization with a planar front [1, 2]. However, such a regime of solidification is actually observed quite rarely. The planar front is usually broken due to thermal or concentration supercooling [3–7]. Concentration supercooling appears ahead of the crystallization front when the concentration gradient exceeds its temperature analogue and creates favorable conditions for the growth of individual protrusions of the solid phase, which produce an extended region of dendrite growth in front of the interface [8].

The problem of selection of stable growth conditions for a dendrite in the melt appeared from analysis of Ivantsov solutions [9–11] and experimental tests for a needle-shaped parabolic crystal [12–18]. Comparative analysis and tests lead to the conclusion that the continuous family of isotropic Ivantsov solutions is unstable: a needle-shaped crystal loses its initial parabolic shape in the steady-state growth regime (see [19] for the review). For this reason, the Ivantsov solution is used as the zeroth approximation in analysis of stable growth in which the surface tension anisotropy parameter or the growth kinetic anisotropy parameter plays the role of the small parameter [20]. After the formulation of the criterion of stable growth of the dendrite tip in a one-component stationary medium [19, 20], the problem was extended to the case of convective flow of the medium [21–23], to the case of dendrite growth in a binary system without convection [24], and to the case of dendrite growth in a binary sys-

tem with convection [25, 26]. However, in many situations a comparative analysis of the dendrite growth taking into account the multicomponent nature of the system is required [27].

In this study, which is a continuation and extension of the above-mentioned works, we analyze the effect of the multicomponent nature of the melt with allowance for the nonlinearity of the liquidus equation [28, 29] on the selection of the steady-state motion of the tip of a parabolic dendrite.

1. FORMULATION OF THE PROBLEM

The growth of a crystal with the phase transition boundary moving into the melt is described by a nonlinear thermodiffusion model of the Stefan type. Temperature T_{int} of the crystal–liquid interface for a one-component system depends on its local curvature $1/R$, crystallization temperature T_0 of the substance, surface tension σ , and latent heat of crystallization Q :

$$T_{\text{int}} = T_0 - \frac{\sigma T_0}{QR}. \quad (1)$$

The temperature field in the solid (T_s) and liquid (T_l) phases of the system is described by the classical heat conduction equations

$$\frac{\partial T_s}{\partial t} = D_T \Delta T_s, \quad \frac{\partial T_l}{\partial t} = D_T \Delta T_l, \quad (2)$$

where D_T is the thermal diffusivity, t is the time, and Δ is the Laplace operator. To simplify the analysis of the problem, we first consider the case of a three-component system and then generalize it to the multicomponent melt. For concentrations C_i and B_i of impurities dissolved in the liquid, we have the diffusion equations [30–32]

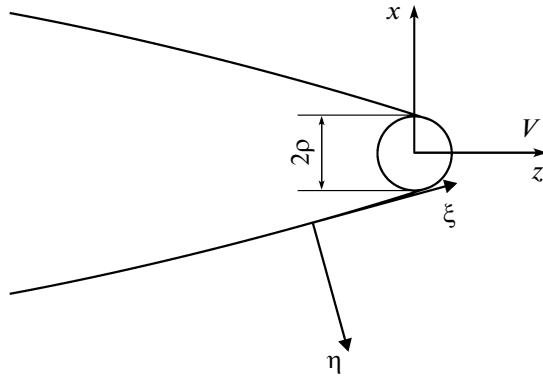


Fig. 1. Schematic diagram of dendrite growth.

$$\frac{\partial C_l}{\partial t} = D_C \Delta C_l, \quad \frac{\partial B_l}{\partial t} = D_B \Delta B_l, \quad (3)$$

where D_C and D_B are the diffusion coefficients for the impurities.

It should specially be noted that in a more rigorous description of diffusion in a multicomponent melt, Fick's equations (3) should be generally replaced by the Onsager equations [32, 33]. Simpler relations (3) hold when supercooling in the system is small [32] and the difference between the concentrations of impurities in the melt in the vicinity of the interface and away from it is on the order of 20% [33]. Upon an increase in supercooling and the corresponding increase in the dendrite growth rate, we must also take into account the local deviations from equilibrium in the crystallization process, which leads to hyperbolic mass transfer equations [34]. Bearing this in mind, we will use simpler relations (3) that are often employed for describing the mass transfer in multicomponent systems (see, for example, [30–32]) to simplify the theory developed here.

The following conditions of the equality of the surface temperature to the phase-transition temperature, of the continuity of temperature, and the heat and mass balance conditions hold at the moving interface:

$$T_l = T_{\text{int}} + F(C_l, B_l), \quad T_s = T_l, \quad (4)$$

$$Q \mathbf{v} \mathbf{n} = D_T c_p (\nabla T_s - \nabla T_l) \mathbf{n},$$

$$(1 - k_C) C_l \mathbf{v} \mathbf{n} + D_C \nabla C_l \mathbf{n} = 0, \quad (5)$$

$$(1 - k_B) B_l \mathbf{v} \mathbf{n} + D_B \nabla B_l \mathbf{n} = 0,$$

where $\mathbf{v} \mathbf{n}$ is the normal velocity of the surface; c_p is the specific heat capacity; k_C and k_B are the equilibrium coefficients of the distribution of impurity of the relevant components; and $F(C_l, B_l)$ expresses in the general case the arbitrary dependence of the phase-transition temperature on impurity concentrations C_l and B_l . Note that in the case of a linear phase diagram of the system, this function can be expressed in terms of con-

stant slopes m and n of the liquidus plane; i.e., $F(C_l, B_l) = -mC_l - nB_l$ [30].

2. ANALYTIC SOLUTION

We assume that a parabolic dendrite grows with a constant rate V along the spatial z axis (Fig. 1). We introduce parabolic coordinates ξ and η connected with Cartesian coordinates x and z by the following relations:

$$x = \rho \sqrt{\xi \eta}, \quad z = \frac{\rho}{2} (\eta - \xi), \quad (6)$$

where ρ is the radius of curvature of the dendrite tip and the interface is at the level $\eta = 1$.

Defining the solution to the problem, which depends on variable η alone, we can write the solutions to Eqs. (2) and (3) in parabolic coordinates (6) in the form (see also [25, 26])

$$\begin{aligned} T_l(\eta) &= T_i + (T_\infty - T_i) \frac{I(\eta)}{I(\infty)}, \\ C_l(\eta) &= C_i + (C_\infty - C_i) \frac{I_1(\eta)}{I_1(\infty)}, \\ B_l(\eta) &= B_i + (B_\infty - B_i) \frac{I_2(\eta)}{I_2(\infty)}, \end{aligned} \quad (7)$$

where C_i , B_i , and T_i are the concentrations of the impurities and the temperature at the interface. In expressions (7), the following notation has been introduced:

$$T_i = T_\infty + \frac{Q}{c_p} P_g \exp(P_g) I(\infty),$$

$$I(\eta) = \int_1^\eta \exp[-P_g \eta'] \frac{d\eta'}{\sqrt{\eta'}}, \quad P_g = \frac{\rho V}{2 D_T},$$

$$C_i = \frac{C_\infty}{1 - (1 - k_C) \exp[P_g D_T / D_C] I_1(\infty) P_g D_T / D_C},$$

$$I_1(\eta) = \int_1^\eta \exp\left[-P_g \frac{D_T}{D_C} \eta'\right] \frac{d\eta'}{\sqrt{\eta'}},$$

$$B_i = \frac{B_\infty}{1 - (1 - k_B) \exp[P_g D_T / D_B] I_2(\infty) P_g D_T / D_B},$$

$$I_2(\eta) = \int_1^\eta \exp\left[-P_g \frac{D_T}{D_B} \eta'\right] \frac{d\eta'}{\sqrt{\eta'}}.$$

Here, P_g is the Peclet growth number and T_∞ , C_∞ , and B_∞ are the temperature and impurity concentrations in the liquid far away from the interface.

3. MICROSCOPIC SOLVABILITY CONDITION

In the case of weak surface tension effects, the solutions with a constant dendrite growth rate can be

found in the vicinity of the classical solutions of the Ivantsov parabolic dendrite if the microscopic solvability condition holds. This allows us to select a stable regime of dendrite growth in terms of velocity V of its tip with radius of curvature ρ (i.e., with superimposed symmetry of the crystal lattice taking into account anisotropy of the preferred direction of crystal growth) [19, 20]. Further, we will use the microscopic solvability condition obtained in [35], which can be written in the form (i is the imaginary unity)

$$\int_{-\infty}^{\infty} G[X_0(l)] Y_m(l) dl = 0, \quad (8)$$

$$Y_m(l) = \exp \left[i \int_0^l k_m(l_1) dl_1 \right].$$

This expression can be used for analyzing the shape of movable boundaries of the frontal type (e.g., Saffman–Taylor “viscous fingers” [19, 36]). For this purpose, we must know the curvature operator G and solutions $X_0(l)$ that give functions $k_m(l)$ of the local non-zero marginal mode of the conjugate dispersion equation for perturbations (see, for example, [36]). Expression (8) can be derived using the Wentzel–Kramers–Brillouin (WKB) method [37] that was used for determining the regimes of propagation of flame fronts [38] and dendrite structure [39]. We will determine below the elements $Y_m(l)$ appearing in relation (9) for the dendrite growth in a three-component melt.

4. LINEAR ANALYSIS OF STABILITY

We will use the results of linear analysis of stability from [23, 25, 26], where the growth rate of perturbations is characterized by a wavelength much smaller than the characteristic spatial scale of the unperturbed solution. We introduce new local Cartesian coordinates (x_c, y_c) associated with the crystal. Here, x_c and y_c denote the axes tangential and normal to the interface, respectively, at the point at which the normal to the surface forms angle θ with the growth axis. Expressing the derivatives of temperature and concentration with respect to coordinates from Eqs. (4) and (5),

$$\frac{\partial T_l}{\partial y_c} = \frac{Q \bar{v}}{D_T c_p}, \quad \frac{\partial C_l}{\partial y_c} = \frac{C_i(1-k_c) \bar{v}}{D_c}, \quad (9)$$

$$\frac{\partial B_l}{\partial y_c} = \frac{B_i(1-k_B) \bar{v}}{D_B}, \quad y_c = 0,$$

we determine the expansions of the temperature and impurity concentrations in the vicinity of the dendrite tip:

$$\bar{T}_l = T_0 - \frac{QV}{D_T c_p} \cos \theta y_c,$$

$$\bar{C}_l = C_i - \frac{C_i(1-k_c)V}{D_c} \cos \theta y_c, \quad (10)$$

$$\bar{B}_l = B_i - \frac{B_i(1-k_B)V}{D_B} \cos \theta y_c, \quad \bar{v} = -V \cos \theta.$$

We denote by ξ' and T'_s the perturbations of the stationary interfacial surface with wavelength λ (which is much smaller than radius ρ of the dendrite tip) and of the temperature field in the solid phase, which can be written in the form [23, 25, 26]

$$\xi' = \Sigma \exp(\omega t + i k x_c - \varepsilon k y_c), \quad (11)$$

$$T'_s = T_{s0} \exp(\omega t + i k x_c - \varepsilon k y_c).$$

Here, Σ and T_{s0} are the amplitudes of perturbations of the dendrite surface and temperature in the solid phase, ω and k are the frequency and the wavenumber of perturbations, respectively, and parameter ε has the same sign as the real part of k because perturbations cannot increase unlimitedly for $y_c \rightarrow +\infty$.

Let us now consider the equation for temperature perturbations in the melt. Retaining only the linear terms, we obtain from Eq. (2)

$$\frac{\partial T'_l}{\partial t} + \bar{u} \frac{\partial T'_l}{\partial x_c} + \frac{\partial T'_l}{\partial y_c} - \frac{\partial \xi'}{\partial t} \frac{\partial \bar{T}_l}{\partial y_c} = D_T \left(\frac{\partial^2 T'_l}{\partial x_c^2} + \frac{\partial^2 T'_l}{\partial y_c^2} \right), \quad (12)$$

$$\bar{u} = -V \sin \theta.$$

Analogously to the theory developed in [23, 25, 26] for binary systems, we will seek the solution to Eq. (12) in the form

$$T'_l = g(y_c) \exp(\omega t + i k x_c - \varepsilon k y_c). \quad (13)$$

Substituting this relation into Eq. (12), we obtain the following equation for amplitude $g(y_c)$:

$$\frac{d^2 g}{dy_c^2} - 2\varepsilon k \frac{dg}{dy_c} = L(g(y_c), y_c), \quad (14)$$

$$L = [\omega + k V \varepsilon \exp(-i\varepsilon\theta)] \frac{g(y_c)}{D_T} + \frac{\omega Q V \cos \theta \Sigma}{c_p D_T^2}.$$

The solution to this equation will be sought in the vicinity of the Mullins–Sekerka solution [15, 35, 36] with constant amplitude $g(y_c) = \bar{T}_{l0} = \text{const}$. Substituting T_{l0} into the right-hand side of the first expression in (14), we obtain the first approximation for $g(y_c)$:

$$g(y_c) = T_{l0} - \left[\left(\frac{\omega}{2\varepsilon k} + \frac{V}{2} \exp(-i\varepsilon\theta) \right) \times \frac{T_{l0}}{D_T} + \frac{\omega Q V \cos \theta \Sigma}{2c_p D_T^2 \varepsilon k} \right] y_c. \quad (15)$$

In this expression, strong inequality $V/D_T \ll k$ is taken into account (here, k is estimated in accordance with the Mullins–Sekerka theory as 10^7 m^{-1} [15, 35, 36])

and V/D_T is estimated as 10^2 m^{-1} for binary metallic systems).

Equations (3) written for perturbations C'_l and B'_l of the concentrations of impurities in the liquid can be solved analogously:

$$\begin{aligned} C'_l &= h_1(y_c) \exp(\omega t + ikx_c - \varepsilon ky_c), \\ B'_l &= h_2(y_c) \exp(\omega t + ikx_c - \varepsilon ky_c), \\ h_1(y_c) &= C_{l0} + \left\{ [\omega + k\varepsilon V \exp(-i\varepsilon\theta)] \frac{C_{l0}}{D_C} \right. \\ &\quad \left. + \frac{\omega C_i(1 - k_C)V \cos \theta}{D_C^2} \Sigma \right\} \frac{y_c}{V \cos \theta / D_C - 2\varepsilon k}, \\ h_2(y_c) &= B_{l0} + \left\{ [\omega + k\varepsilon V \exp(-i\varepsilon\theta)] \frac{B_{l0}}{D_B} \right. \\ &\quad \left. + \frac{\omega B_i(1 - k_B)V \cos \theta}{D_B^2} \Sigma \right\} \frac{y_c}{V \cos \theta / D_B - 2\varepsilon k}. \end{aligned} \quad (16)$$

Perturbing boundary conditions (4) and (5), we obtain the following relations at the interface $y_c = 0$:

$$\begin{aligned} T'_l &= \frac{QV \cos \theta}{D_T c_p} \xi' - m C'_l - n B'_l + \left(\frac{m C_i(1 - k_C)V \cos \theta}{D_C} \right. \\ &\quad \left. + \frac{n B_i(1 - k_B)V \cos \theta}{D_B} \right) \xi' - \frac{Qd \partial^2 \xi'}{c_p \partial y_c^2}, \\ T'_s &= m C'_l + n B'_l - \left(\frac{m C_i(1 - k_C)V \cos \theta}{D_C} \right. \\ &\quad \left. + \frac{n B_i(1 - k_B)V \cos \theta}{D_B} \right) \xi' + \frac{Qd \partial^2 \xi'}{c_p \partial y_c^2}, \\ \frac{Q}{D_T c_p} \frac{\partial \xi'}{\partial t} &= \frac{\partial T'_s}{\partial y_c} - \frac{\partial T'_l}{\partial y_c} - \frac{QV^2 \cos^2 \theta}{D_T^2 c_p} \xi', \\ \frac{1 - k_C}{D_C} \left(C'_l \frac{\partial \xi'}{\partial t} + V \cos \theta C'_l \right) &+ \frac{\partial C'_l}{\partial y_c} \\ &+ \frac{C_i k_C(1 - k_C)V^2 \cos^2 \theta}{D_C^2} \xi' = 0, \\ \frac{1 - k_B}{D_B} \left(B'_l \frac{\partial \xi'}{\partial t} + V \cos \theta B'_l \right) &+ \frac{\partial B'_l}{\partial y_c} \\ &+ \frac{B_i k_B(1 - k_B)V^2 \cos^2 \theta}{D_B^2} \xi' = 0, \end{aligned} \quad (17)$$

where $d = \sigma c_p T_0 / Q^2$ denotes the capillary length, and $m = -\partial F / \partial C_i$ and $n = -\partial F / \partial B_i$ are the derivatives calculated on the dendrite surface for $C_l = C_i$ and $B_l = B_i$. Substituting perturbations (11), (13), and (16) into

system (17), we obtain five equations for amplitudes Σ , T_{l0} , T_{s0} , C_{l0} , and B_{l0} . The zero value of the determinant of this system, which is composed for the coefficients of the perturbation amplitudes, defines the equation for wavenumber k . In the coordinate system with the origin moving in the direction of the normal to the interface with velocity $V \cos \theta$, the perturbation with wavenumber k increases at a rate $\omega(k)$ with increasing wavenumber k on account of the rotational symmetry of the system. If the origin moves now in the direction of the z axis with constant velocity V , the perturbation growth rate is $\omega(k) + iV k \sin \theta$ owing to the tangential velocity $V \sin \theta$ of the new system of coordinates [21]. Therefore, substituting $-iV k \sin \theta$ for $\omega(k)$, we obtain the following equation for the wavenumber on the neutral stability curve for $\omega = 0$:

$$\begin{aligned} k &= k_{TC} \sqrt{\frac{\exp(i\theta)}{1 - \beta \cos 4\theta}}, \quad k_{TC} = -\sqrt{\frac{VP}{2d_0 D_T}}, \\ P &= 1 + 2 \frac{m C_i(1 - k_C) D_T c_p}{Q D_C} + 2 \frac{n B_i(1 - k_B) D_T c_p}{Q D_B}. \end{aligned} \quad (18)$$

In these expressions, $k \sim 10^7 \text{ m}^{-1}$, $V/D_T \sim 10^2 \text{ m}^{-1}$, $V/D_C \sim 10^6 \text{ m}^{-1}$, $V/D_B \sim 10^6 \text{ m}^{-1}$, $d \sim 10^{-10} \text{ m}$, $\rho \sim 10^{-5} \text{ m}$ according to estimates, and only the terms corresponding to the solution for the one-component, binary, and three-component systems are taken into account. Writing relations (18), we have also taken into account the expression for capillary length $d(\theta) = d_0(1 - \beta \cos 4\theta)$ and the smallness of angle θ , where $\beta = 15\varepsilon_c \ll 1$ is the anisotropy factor, ε_c is the anisotropy parameter of the surface energy at the interface, and d_0 is the capillary constant [23, 27].

It should be noted that wavenumber (18) for $C_i = 0$ ($D_C \rightarrow \infty$) and $B_i = 0$ ($D_B \rightarrow \infty$) is transformed to the corresponding Mullins–Sekerka expression for the one-component system [15, 35, 36]. Expression (18) also contains a correct limiting transition to the wavenumber for the binary system [24] for $C_i = 0$ ($D_C \rightarrow \infty$) and $B_i = 0$ ($D_B \rightarrow \infty$). Upon an increase in the impurity concentration of the third component, the critical wavenumber increases. Thus, relation (18) gives the critical wavenumber for perturbations at the dendrite tip in the three-component thermoconcentration problem. In the case of a large number of dissolved components, the expression for parameter P should obviously be supplemented with the corresponding terms.

5. CRITERION OF STABLE GROWTH OF THE DENDRITE TIP

To formulate the stability criterion, we can use the microscopic solvability condition (8). Considering that

$$l = \frac{-\rho}{2} \left[\frac{\tan \theta}{\cos \theta} + \ln \left(\frac{1}{\cos \theta} + \tan \theta \right) \right]$$

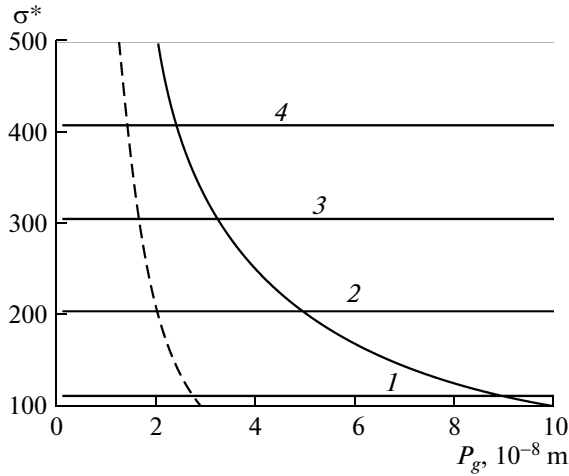


Fig. 2. Scaling factor as a function of the Peclet number $P_g = \rho V / 2D_T$ in accordance with relation (21) for a fixed radius of curvature ρ of the dendrite tip (solid curve, $d_0/\rho = 10^{-5}$) and a fixed growth rate V (dashed curve, $Vd_0/D_T = 1.7 \times 10^{-13}$) as functions of impurity concentration $B_\infty = 0.1$ (1), 1 (2), 2 (3), and 3 at % (4) for $k_B = 0.5$. The values of the computation parameters of the system are [23, 25, 26]: $D_T/D_C = D_T/D_B = 5 \times 10^3$, $k_C = 0.5$, $C_\infty = 1$ at %, $m = n = 10^\circ\text{C/at \%}$, $Q/c_p = 300^\circ\text{C}$, $\beta = 0.75$, and $\sigma_0 = 1$.

(see, for example, [35]) and following [23], we can write solvability condition (8) in the form

$$\int_{-\infty}^{+\infty} G(\chi) \exp[\sqrt{G}\psi(\chi)] d\chi = 0, \quad \chi = \tan\theta, \quad (19)$$

$$\psi(\chi) = \frac{i}{2} \int_0^\chi \frac{(1+i\chi')(1+\chi'^2)^{5/2}}{B(\chi')} d\chi',$$

$$B(\chi) = (1-\beta)(1+\chi^2)^2 + 8\beta\chi^2,$$

where constant C is normalized to dimensionless factor $VP\rho^2/(2d_0D_T)$.

Further, we estimate integral (19) in the weak anisotropy limit using the method developed in [23]. The numerator of the integrand vanishes for $\chi = i$ (stationary phase point), while the denominator vanishes for $\chi = i(1 - \sqrt{2\beta})$ (singularity point). Since the main contribution to the integral comes from the neighborhood of point $\chi = i$ [23], function $\psi(\chi)$ can be approximated by the expression

$$\psi(\chi) = 2^{9/8} \beta^{7/8} \int_{1/\sqrt{2\beta}}^\phi \frac{\phi'^{7/4} d\phi'}{\sqrt{\phi'^2 - 1}}, \quad (20)$$

$$\chi = i(1 - \sqrt{2\beta}\phi).$$

The integral in this formula can be evaluated using the approximate methods developed in [23] in analysis of the problem of the dendrite growth in a one-component system with a convective flow. Following this method, note that two main contributions come to

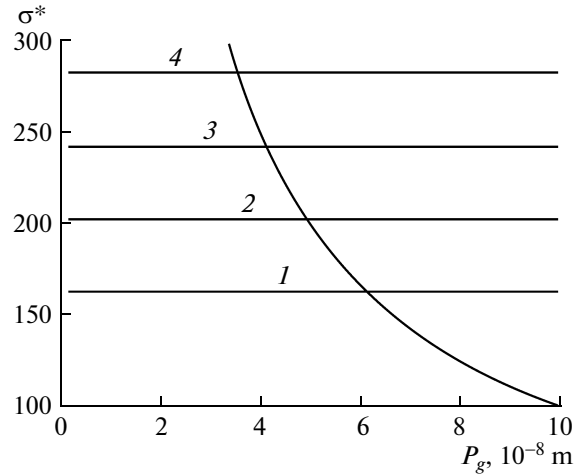


Fig. 3. Scaling factor as a function of the Peclet number $P_g = \rho V / 2D_T$ in accordance with relation (21) for a fixed radius of curvature of the dendrite tip as a function of the impurity distribution coefficient $k_B = 0.7$ (1), 0.5 (2), 0.3 (3), and 0.1 (4) for $B_\infty = 1$ at %. The values of computation parameters of the system are the same as in Fig. 2.

integral (20): the contribution from the integration over the loop and that from stationary phase points. The first contribution is calculated at distance $\phi' \sim 0$, which results in the oscillating factor of the exponentially small integral $\cos(A_1 \sqrt{C} \beta^{7/8})$. Each stationary phase point makes a contribution with the oscillating part of the form $\cos(A_2 \sqrt{C} \beta^{7/8})$, where A_1 and A_2 are constants. Neutralization of the sum of these contributions defines quantity $C = q^2/\beta^{7/4}$, where q is an integer. The allowance for the normalization in relation (20) now leads to the following expression for scaling factor σ^* in the form

$$\sigma^* \equiv \frac{2d_0D_T}{\rho^2V} = \sigma_0 \beta^{7/4} \left(1 + 2 \frac{D_T c_p m C_i (1 - k_C)}{Q D_C} + 2 \frac{D_T c_p n B_i (1 - k_B)}{Q D_B} \right), \quad (21)$$

where σ_0 is the numerical constant defined with the help of asymptotic methods [20] or by combining model predictions with experimental values [40, 41].

CONCLUSIONS

Expression (21) is the central result of the theory being developed. It defines the criterion for the stable mode of the dendrite tip growth in a three-component melt taking into account the surface energy anisotropy (parameter β) and the nonisothermal nature of the liquid. Criterion (21) is a generalization of the results obtained earlier for one-component [15, 35, 36] and binary [24] melts disregarding convection.

Figures 2 and 3 show the dependences of both parts of expression (21) on the Peclet growth number. It can

easily be seen that an increase in impurity concentration B_∞ of the third component, as well as a decrease in its distribution coefficient k_B (an increase in the fraction of the impurity expelled in front of a dendrite), reduces the number of selected values of the Peclet number, solidification rate V , and radius of curvature ρ of the dendrite tip, which are determined by the points of intersection of hyperbolas with corresponding lines in the figures. In other words, accumulation of the impurity in front of a dendrite makes its tip thinner and slows down its growth rate.

Criterion (21) can easily be generalized to the case of the multicomponent system:

$$\sigma^* \equiv \frac{2d_0 D_T}{\rho^2 V} \quad (22)$$

$$= \sigma_0 \beta^{7/4} \left(1 + 2 \frac{D_T c_p}{Q} \sum_{j=1}^N \frac{m_j C_j (1 - k_j)}{D_j} \right),$$

where N is the number of the mixture components, $m_j = -\partial F / \partial C_j$, $F = F(C_1, C_2, \dots)$, and C_j , k_j , and D_j are the concentrations of the impurity, the coefficients of its distribution, and the diffusion coefficients for the j th component. A number of important features of the theory being developed are also worth noting.

Criteria (21) and (22) formulated above for the stable growth of a dendrite in a multicomponent system can be verified, for example, by computer simulation methods (e.g., the phase field method employed earlier for studying the free growth of a dendrite in a one-component system with convection [42, 43]). The stability criteria formulated here can also be verified experimentally as, for example, in the case of dendrite growth in a forced flow of a transparent fluid [44].

The theory developed here, as well as the theories constructed in earlier publications [23–26, 35, 36], is limited to analysis of relatively small values of the Peclet growth number $P_g = \rho V / 2D_T$. In other words, the theory is valid only for small gradients or low supercooling of the fluid, which ensure low rates of dendrite growth. An extended analysis of elevated growth rates for arbitrary Peclet numbers requires special investigation of stability of high-rate regimes of dendrite solidification. The derivation and analysis of the microscopic solvability condition for a high-rate locally nonequilibrium regime of growth can be performed in accordance with the theory developed in [45–47].

Criteria (18), (21), and (22) formulated in this study for a 2D dendrite will also hold in the 3D case because these criteria are independent of the spatial dimensionality of the problem. For a 3D dendrite, the corresponding solution to the thermodiffusion problem obtained in [48] should be used instead of expressions (7). In this case, other boundary conditions for the impurity concentrations on the dendrite surface, which are determined by the solution of the 3D prob-

lem [48], must be simply substituted into relations (18), (21), and (22).

ACKNOWLEDGMENTS

This study was financed in part by the Russian Foundation for Basic Research (project nos. 11-01-00137 and 10-01-96045-Ural) and the Federal Target Program “Scientific and Academic—Teaching Staff of Innovative Russia” in 2009–2013.

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Translated by N. Wadhwa